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Offset susceptibility of superconductors

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The novel concept of offset susceptibility χ_0 is presented for superconductors in applied fields $H_{dc}+H_{ac}\sin\omega t$. χ_0 represents the dc component of the time-dependent magnetization $M(t)$. For sintered $YBa_2Cu_3O_7$, χ_0 was measured as a function of H_{ac}, H_{dc}, and temperature using a Hall probe magnetometer and Fourier analysis. For *intra*granular diamagnetism, χ_0 behaves as the real part of complex fundamental susceptibility χ_1', confirming a London-like field penetration. However, χ_0 arising from *inter*granular coupling is *positive* at temperatures just below the onset of the coupling transition, in good agreement with theoretical predictions of a simplified Kim critical-state model of magnetization.

A sintered high-T_c superconductor, such as $YBa_2Cu_3O_7$, can be regarded as an assembly of weakly connected superconducting grains. The magnetic response of such materials is very useful in their characterization. After a sample is cooled in zero field, the dc susceptibility χ_{dc} typically shows a two-step structure as a function of temperature, and the complex fundamental susceptibility $\chi_1 \equiv \chi_1' - i\chi_1''$ has corresponding intrinsic (*intra*granular) and coupling (*inter*granular) components. For a time-dependent magnetic excitation field, $H(t)=H_{dc}+H_{ac}\sin\omega t$, at the fundamental frequency ω ($\equiv 2\pi f$), the coupling component is very sensitive to temperature T near T_c, the amplitude of the ac magnetic field H_{ac}, and the superimposed dc field H_{dc}. In addition, harmonic susceptibilities $\chi_n \equiv \chi_n' - i\chi_n''$ (n =integer) appear below T_c.[1-4]

A successful explanation for odd and even complex harmonic susceptibilities of high-T_c superconductors is a Kim critical-state model,[5] which assumes a local reciprocal magnetic-field dependence of the critical current density J_c in a weak-field region on the order of 1 mT.[2,3] Such a field dependence of J_c may be expected if J_c is determined by an average of the Josephson critical current diffraction patterns of various periods as functions of H.[4] In this sense, weak links dominate the intergranular electrodynamics of sintered high-T_c superconductors, and are responsible for the appearance of the even harmonics. On the other hand, a two-dimensional numerical simulation of a Josephson network also seems to describe the odd and even harmonics.[6]

Often the ac and dc susceptibilities of superconductors are treated independently. The dc susceptibility χ_{dc} is simply defined by the ratio M/H_{dc} using the volume magnetization M and the external field H_{dc}. In this paper, we extend the definition of χ_{dc} for excitation fields $H(t)=H_{dc}+H_{ac}\sin\omega t$, which results in a novel offset susceptibility χ_0. Experimental and theoretical χ_0 data are presented to examine intergranular properties of sintered $YBa_2Cu_3O_7$.

The real part of complex susceptibility χ_1' represents the dispersive magnetic response, and the imaginary part χ_1'' represents energy dissipation. For a nonlinear magnetic system, we expand the volume magnetization $M(t)$ for an external magnetic field $H(t)=H_{dc}+H_{ac}\sin\omega t$ as

$$M(t)=\chi_0 H_{dc}+H_{ac}\sum_{n=1}^{\infty}[\chi_n' \sin(n\omega t)-\chi_n'' \cos(n\omega t)] , \quad (1)$$

where $\chi_0 H_{dc}$ is a dc *offset* component and the complex harmonic susceptibilities $\chi_n \equiv \chi_n' - i\chi_n''$ ($n=1,2,3,\ldots$) are defined as the Fourier coefficients in Eq. (1). [In Refs. 1 and 2, an alternative definition of harmonic susceptibility is given for a field defined as $H(t)=H_{dc}+H_{ac}\cos\omega t$.] We obtain the dc offset susceptibility χ_0 by

$$\chi_0=\frac{1}{2\pi H_{dc}}\int_0^{2\pi} M(t)\,d(\omega t) , \quad (2)$$

where $H_{dc}\neq 0$. The offset susceptibility χ_0 coincides with the dc susceptibility χ_{dc} in the limit $H_{ac}\to 0$.

With a conventional ac susceptometer, such as that used in Ref. 2, a voltage $v(t)\propto dM(t)/dt$ is detected; such a susceptometer is unable to detect any dc component of $M(t)$. To measure χ_0, we used a cryogenic Hall probe to directly sense the sample magnetization $M(t)$ as a function of time. (A vibrating-sample magnetometer could also be used for this purpose.) The sample was initially cooled in zero field before each measurement to avoid any effects associated with trapped vortices. Temperature of the sample was measured with a Pt resistance thermometer. At 76 K, χ_0 was measured as a function of increasing H_{ac} or increasing H_{dc}. Also χ_0 was measured as a function of increasing T at constant H_{dc} and H_{ac}. The current to the field coil was generated by a constant-current amplifier driven by a 14-bit digital-analog voltage converter. The wave form $H(t)$ for a single measurement was given by $H_{dc}t/\tau$ for $0\le t<\tau$, $H_{dc}+H_{ac}\sin 2\pi f t$ for $\tau\le t<3\tau$, and $H_{dc}(4-t/\tau)$ for $3\tau\le t<4\tau$. We fixed the period τ as 1024×2 ms

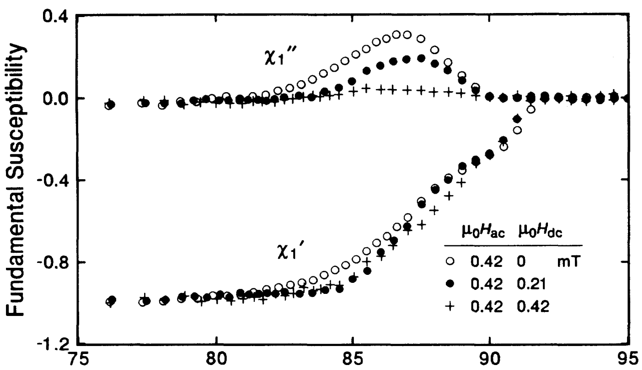

FIG. 1. Complex fundamental susceptibility χ_1' and χ_1'' of sintered $YBa_2Cu_3O_7$ as a function of T measured with a Hall probe magnetometer ($\mu_0 H_{ac}=0.42$ mT, $\mu_0 H_{dc}=0$, 0.21, 0.42 mT). Theoretical curves are available in Ref. 2.

($f=\tau^{-1}=0.488$ Hz) for all measurements. An $YBa_2Cu_3O_7$ sample ($10.7\times1.2\times1.0$ mm) was prepared by a standard solid-state reaction at 905 °C for 10 h. The field was applied perpendicular to the 10.7×1.2-mm surface of the sample, giving a demagnetizing factor of ~ 0.5. The axial Hall probe was glued to the sample, aligned so as to be insensitive to the external field $H(t)$, but sensitive to the nonaxial component of the field from the sample magnetization $M(t)$.[7] The sensor was calibrated based on the perfect diamagnetic shielding of the superconducting sample at 76 K. The output of a Hall gaussmeter was amplified by a low-noise preamplifier with a low-pass filter. We chose a cutoff frequency of 30 Hz for the filter to reduce noise. [This causes a phase shift of 1.8° in $M(t)$ at $f\simeq 0.5$ Hz.] We sampled 4096 points for both $H(t)$ and $M(t)$ using a 16-bit analog-digital converter as a function of t $(0\le t<4\tau)$, corresponding to a phase resolution of 0.35°. For analysis, we took 1024 points in the period $2\tau\le t<3\tau$. The first cycle $(\tau\le t<2\tau)$ cancels all memory of prior flux profiles because the measurements are carried out with increasing H_{ac}, H_{dc}, or T. We calculated χ_0 and $\chi_n'-i\chi_n''$ $(n\ge1)$ from $M(t)$ using fast Fourier transforms. We also synthesized the M-H magnetization hysteresis loop from $M(t)$ and $H(t)$, where time t is a parameter. [Note that the inverse procedure, the reconstruction of the M-H curve from the measured χ_n's using Eq. (1), *requires* $\chi_0 H_{dc}$.]

Various models are available to obtain theoretical $M(t)$ of granular superconductors. The dc offset susceptibility χ_0 can then be obtained from $M(t)$ using Eq. (2). The function $M(t)$ consists of the appropriate terms for magnetization as a function of field $M(H)$, where H appears as $H(t)=H_{dc}+H_{ac}\sin(\omega t)$.[3] Otherwise, one could obtain the discrete values of $M(t_i)$ numerically or by simulation at t_i, $0\le t_i<2\pi/\omega$, and use fast Fourier transforms or numerical integrations to obtain χ_0 and χ_n $(n\ge1)$.

The Bean and Ishida-Mazaki models for magnetization do not predict either offset or even harmonic susceptibilities.[8,9] To account for nonzero χ_0 and even harmonics, it is sufficient to introduce a local-field H_i dependence of critical current density J_c. Kim, Hempstead, and Strnad[5] assumed $J_c(H_i)=k/(H_0+|H_i|)$, where k and H_0 are positive constants. Ji *et al.*[3] derived equations of sample magnetization $M(H)$ for an infinite slab of thickness $2a$ and a field $H_{dc}+H_{ac}\sin(\omega t)$ using a simplified Kim model, $J_c(H_i)=k/|H_i|$, where the field for full flux penetration H_p is given by $(2ka)^{1/2}$. With a dc offset field, the magnetization curve becomes asymmetric for positive and negative half periods of the ac field $[M(t+\pi/\omega)\neq -M(t)]$. This corresponds to the appearance of even harmonics. Fundamental, harmonic, and dc offset susceptibilities are described by Fourier formulas, but analytic expressions for harmonic susceptibilities have not been obtained for the simplified Kim model. (Such analytic formulas have been given in the limit $H_{dc}\gg H_{ac}$.[1]) We can numerically obtain χ_0, χ_n', and χ_n'' using Ji's equations for various combinations of H_{ac}, H_{dc},

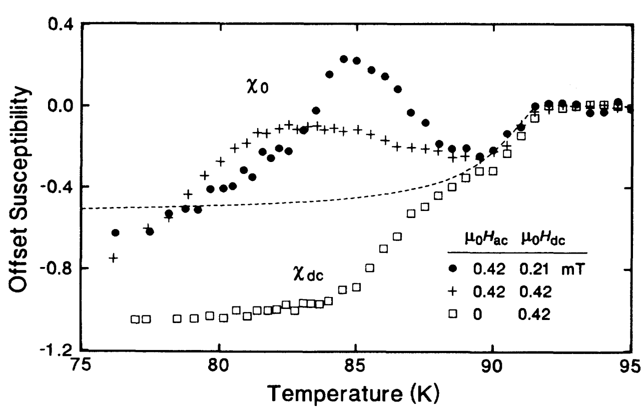

FIG. 2. Offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ pellet as a function of T for three combinations of $\mu_0 H_{ac}$ and $\mu_0 H_{dc}$. The curve taken with $H_{ac}=0$ is equivalent to the zero-field-cooled dc susceptibility χ_{dc}. The dashed curve, representing the intragranular susceptibility, is a guide for the eye.

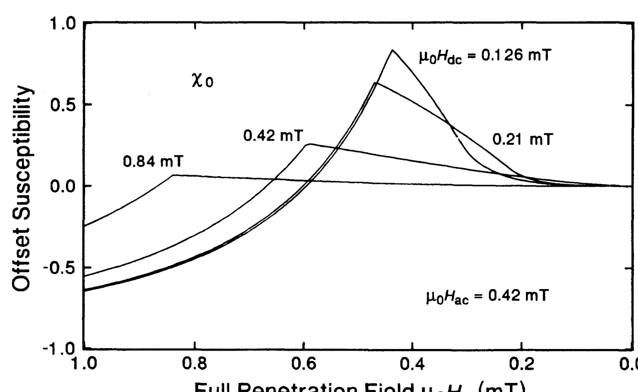

FIG. 3. Theoretical offset susceptibility χ_0 as a function of $\mu_0 H_p$ ($\mu_0 H_{ac}=0.42$ mT, $\mu_0 H_{dc}=0.126$, 0.21, 0.42, 0.84 mT). H_p is inversely related to T. Compare with Fig. 2.

and $H_p(T)$. The simplified Kim model was successful in explaining the intergranular complex harmonic susceptibilities of a sintered high-T_c superconductor,[2] but the examination of χ_0 offers another check of the model.

In Fig. 1, we show χ_1' and χ_1'' for an $YBa_2Cu_3O_7$ sample as functions of T, demonstrating that the Hall probe magnetometer is able to measure the conventional ac susceptibility. [We normalized χ_0, χ_n, and $M(t)$ so as to give -1 for χ_1' at 76 K.] χ_1' as a function of T shows both intragranular and intergranular components. Characteristic features of χ_1' and χ_1'' are very similar to those typical for ac susceptibility obtained with a susceptometer: Upon increasing H_{dc}, χ_1' arising from intergranular coupling initially shifts to *higher* temperatures and the intergranular peak of χ_1'' is significantly suppressed.[2] The negative value of χ_1'' at 76 K arises from the phase shift in the low-pass filter. This could be easily corrected numerically. We also derived the higher harmonic susceptibilities from $M(t)$ by Fourier analysis and found good agreement with those obtained using lock-in detection with a conventional susceptometer.[2]

In Fig. 2, we show the offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ sample as a function of temperature. The χ_0 curve (open squares) obtained in a pure dc field ($H_{ac}=0$ but $H_{dc}\neq 0$) is equivalent to the zero-field-cooled dc susceptibility χ_{dc}, showing a two-step variation as a function of T. The dashed curve is a guide for the eye intended to indicate the intragranular contribution to both χ_{dc} and χ_1'. Upon application of H_{ac} in addition to H_{dc} (closed circle and plus symbols), χ_0 has an exotic profile just below the onset temperature of intragranular coupling. For the intragranular step region near T_c, χ_0 and χ_{dc} seem to show the same behavior as χ_1' of Fig. 1. This indicates a linear magnetic response. The field penetrates into grains as specified by the London equation both for H_{dc} and $H_{ac}\sin\omega t$ components. This is consistent with the absence of an intragranular peak in χ_1'' in Fig. 1.

In Fig. 3, we show numerically calculated values of χ_0 as a function of $\mu_0H_p(T)$ for four values of μ_0H_{dc} at constant μ_0H_{ac}. Since the model does not account for intragranular susceptibility, the experimental χ_0 curve of Fig. 2 should be interpreted by subtracting the contribution represented by the eye-guide curve. Striking is the prediction of *positive* susceptibility just below T_c, in agreement with the experimental χ_0 of Fig. 2. At larger H_p (corresponding to lower temperatures), χ_0 approaches -1. By comparison of the theoretical and experimental curves, we estimate the full-penetration field $\mu_0H_p\sim 1$ mT.

In Fig. 4, we show the offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ sample as a function of μ_0H_{ac} at 76 K for three values of μ_0H_{dc}. A positive peak for χ_0 vs H_{ac} appears at lower fields. In Fig. 5, the theoretical χ_0 curves show reasonable agreement with those in Fig. 4, where we take $\mu_0H_p=0.924$ mT to simulate a fixed temperature 76 K. A slight increase of χ_0 at higher H_{ac} may be due to the field penetration into grains. (The grains may not be entirely ideal but contain some intragranular weak links.) The local M-H curves also showed good agreement with theoretical predictions (see Fig. 2 of Ref. 1) after subtracting an intragranular contribution.

In Fig. 6, we show the offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ sample as a function of H_{dc} at 76 K for three values of H_{ac}. A positive peak for χ_0 vs H_{dc} is seen again. In Fig. 7, we show the theoretical χ_0 for comparison with those in Fig. 6. Again reasonable agreement can be seen. Further improvement of the theoretical curve might be expected by using the complete Kim model, which contains an additional parameter. We note that the theoretical profile of χ_0 vs H_{dc} is very sensitive to the H_p value. If we change H_p by 10%, we obtain very different features in χ_0 vs H_{dc}. χ_0 at $H_{dc}=0$ is not uniquely defined by Eq. (1). However, as suggested in Fig. 7, χ_0 may be deduced for each finite H_{ac} in the limit $H_{dc}\to 0$.

In conclusion, we discussed the concept of offset susceptibility χ_0 and the use of a Hall probe magnetometer to measure χ_0, the ac fundamental susceptibility $\chi_1'-i\chi_1''$, as well as higher harmonic susceptibilities. This method covers a wide range of parameters H_{dc} and H_{ac}. As one

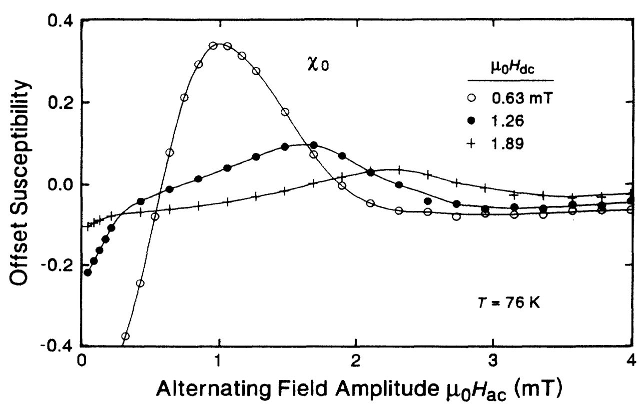

FIG. 4. Offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ sample as a function of μ_0H_{ac} ($T=76$ K, $\mu_0H_{dc}=0.63, 1.26, 1.89$ mT).

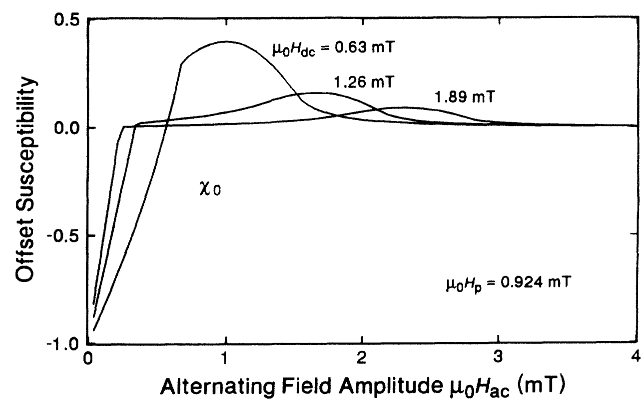

FIG. 5. Theoretical χ_0 as a function of μ_0H_{ac} for three values of μ_0H_{dc}. We take $\mu_0H_p=0.924$ mT to represent $T=76$ K. Compare with Fig. 4.

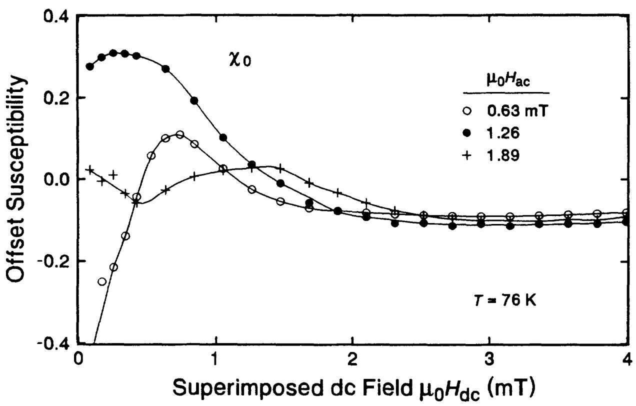

FIG. 6. Offset susceptibility χ_0 of the $YBa_2Cu_3O_7$ sample as a function of $\mu_0 H_{dc}$ ($T=76$ K, $\mu_0 H_{ac}=0.63$, 1.26, 1.89 mT).

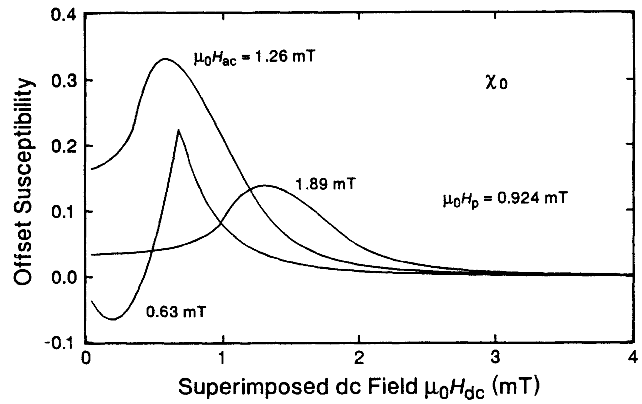

FIG. 7. Theoretical χ_0 as a function of $\mu_0 H_{dc}$ for three values of $\mu_0 H_{ac}$. We take $\mu_0 H_p = 0.924$ mT to represent $T=76$ K. Compare with Fig. 6.

extreme case ($H_{dc} \neq 0$ and $H_{ac}=0$), the dc susceptibility can be measured. As an ac susceptometer, this method is of benefit for very low-frequency applications because the signal magnitude is independent of frequency, in marked contrast with lock-in detection using a pickup coil. We measured χ_0 as a function of H_{ac}, H_{dc}, and T to characterize an $YBa_2Cu_3O_7$ sample. The observed profiles are exotic but in good agreement with the predictions from a simplified Kim critical-state model, giving further confirmation for the validity of the model to describe the intergranular magnetic response for high-T_c superconductors. The offset susceptibility χ_0 could also be used to examine other models with field-dependent J_c, and to verify various models of nonlinear magnetization of magnetic substances such as spin glasses.

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